Light polarization independent nuclear spin alignment in a quantum dot

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We report optical pumping of neutral quantum dots leading to nuclear spin alignment with direction insensitive to polarization and wavelength of light. Measurements of photoluminescence of both "dark" and "bright" excitons in single dots reveal that nuclear spin pumping occurs via a virtual spin-flip transition between these states accompanied by photon emission. The sign of the nuclear spin polarization is determined by asymmetry in the exciton energy spectrum, rather than by the sign of the exciton spin polarization.

Control and understanding of the nuclear spin environment in nano-structures is of great importance in achieving robust coherence of spin-based qubits in the solid state. Recently, optical pumping of nuclear spins in quantum dots (QDs) has been demonstrated [1–8]. Dynamic nuclear polarization (DNP) due to the electron-nuclear hyperfine interaction (HI) occurs when non-equilibrium populations of electron spin states are created using resonant or quasi-resonant circularly polarized light of high intensity. Under such conditions, a direct correspondence between the sign of circular polarization of the exciting light and the direction of the nuclear spin alignment is observed [1–5, 9]. By contrast, suppression of the electron spin alignment under above barrier non-resonant excitation results in negligible nuclear spin polarization in a dot.

Here we report measurements on individual neutral InP/GaInP quantum dots, which shed new light on the mechanisms of DNP in semiconductor nano-structures. They reveal previously unobserved phenomena: at low pumping levels, *independent* of light polarization and wavelength, optical pumping induces an effective nuclear field, which is *always parallel* to the external field applied along the growth axis of the structure. We show that at low power DNP in a neutral dot occurs via the second order recombination of "dark" excitons accompanied by electron-nuclear spin flip-flop. This is revealed in photoluminescence (PL) measurements where optically inactive "dark" excitons are observed due to weak mixing with the "bright" states. Asymmetry in the energy splitting of the excitonic energy levels induced by electron-hole exchange interaction leads to a strong difference of DNP rates induced by "dark" excitons with opposite spins [10]. As a result, the direction of nuclear polarization is independent of the average electron spin polarization on the dot. It is instead controlled by the direction of the external magnetic field experienced by the exciton.

By contrast in the regime of higher powers, where the exciton states of the dot are saturated, we find a direct correspondence between the helicity of light and the direction of the nuclear spin alignment, as was observed previously [1–5, 9]. In this regime DNP is determined by optical orientation of electrons in excited states in the dot or in the wetting layer.

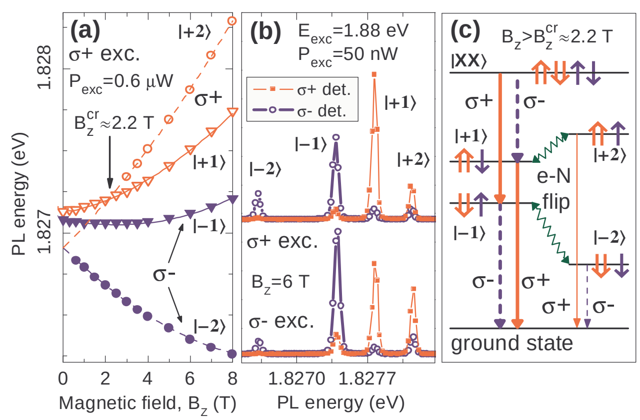

FIG. 1. (a) Magnetic field dependence of exciton PL energies (symbols), lines show fitting. (b) PL spectra of the exciton in a neutral quantum dot detected in σ^+ (■) and σ^- (◯) polarizations under σ^+ (top) and σ^- (bottom) excitation with photon energy 1.88 eV at B_z=6.0 T. (c) Energy diagram of exciton and biexciton states in high magnetic field $B_z > B_z^{cr} \approx 2.2$ T. Heavy holes $\Uparrow(\Downarrow)$ and electrons $\uparrow(\downarrow)$ with spin parallel (antiparallel) to external field form optically allowed ($\Uparrow\downarrow$, $\Downarrow\uparrow$) and "dark" ($\Uparrow\uparrow$, $\Downarrow\downarrow$) excitons with total spin $J_z = \pm 1$ and ± 2 respectively. Thick arrows show circularly polarized recombination of biexciton and "bright" excitons, thin arrows correspond to weak recombination of "dark" exciton states. Zigzag lines show electon-nuclear ($e{-}N$) spin-flips induced by the hyperfine interaction.

The experiments were performed on an undoped InP/GaInP sample without electric gates. PL of neutral InP QDs was measured at T = 4.2 K, in external magnetic field B_z up to 8 T normal to the sample surface. QD PL at ∼1.84 eV was excited with a laser either below (E_{exc}=1.88 eV) or above (E_{exc}=2.28 eV) the GaInP barrier band-gap and analyzed with a 1 m double spectrometer and a CCD.

In a neutral dot electrons $\uparrow(\downarrow)$ and heavy holes $\Uparrow(\Downarrow)$ with spin parallel (antiparallel) to the growth axis Oz can form either optically-forbidden ("dark") excitons $|\Uparrow\uparrow\rangle$

($|\Downarrow\downarrow\rangle$) with spin projection $J_z = +2(-2)$, or "bright" excitons $|\Uparrow\downarrow\rangle$ ($|\Downarrow\uparrow\rangle$) with $J_z = +1(-1)$ optically allowed in $\sigma^+(\sigma^-)$ polarization. The electron-hole ($e-h$) exchange interaction splits off "dark" excitons by the energy δ_0. An additional splitting $\delta_b(\delta_d)$ of the bright(dark) exciton doublet is caused by the reduced symmetry of the QD potential [1, 11]. QD axis misorientation or symmetry reduction may lead to weak mixing of "bright" and "dark" states allowing observation of the latter in PL [11, 12]. Below we denote mixed exciton states with spin projections $J_z \approx \pm 2, \pm 1$ as $|\pm 2\rangle$, $|\pm 1\rangle$ distinguishing them from "pure" excitons $|\Downarrow\uparrow\rangle$, $|\Uparrow\downarrow\rangle$, $|\Uparrow\uparrow\rangle$ and $|\Downarrow\downarrow\rangle$. PL energies of bright (dark) excitons $E_{b(d)}$ measured as functions of B_z in a single neutral dot with $\delta_0 \approx 200$ μeV, $\delta_{b(d)} \approx 65(0)$ μeV are shown in Fig. 1 (a) with triangles (circles). Fitting of the $E_{b(d)}(B_z)$ dependences [lines in Fig.1(a)] allows electron (hole) g-factor $g_{e(h)} \approx +1.6(+2.7)$ to be extracted (see appendix in Ref. [12] for more details on QD characterization).

At high B_z exceeding both the effective exchange field $\delta_b/(\mu_B|g_h - g_e|) \approx$1 T and the field $B_z^{cr} \approx$2.2 T, where $|+1\rangle$ and $|+2\rangle$ states cross, PL originating from all four exciton states can be resolved as shown in Fig. 1 (b) where PL spectra detected in σ^+ (σ^-) polarization at B_z=6.0 T under σ^+ and σ^- polarized excitation are shown by the thin (thick) lines. The energy level structure of all exciton states as well as the biexciton $|\Uparrow\Downarrow\uparrow\downarrow\rangle$ at $B_z > B_z^{cr}$ is shown in Fig. 1 (c) along with possible optical transitions.

Fig. 2 shows PL intensities I^{PL} (left scale) of all four exciton transitions and the total biexciton intensity at B_z=6.0 T as functions of optical power P_{exc} of σ^+ and σ^- polarized excitation with photon energy $E_{exc} = 1.88$ eV. Due to their small oscillator strengths saturation of dark excitons is observed at $P_{exc} \approx 1$ μW, whereas bright excitons and biexcitons saturate at much higher powers of 20 μW and 40 μW, respectively.

The net nuclear field B_N along Oz acting via the HI shifts exciton states with electron spin $\uparrow(\downarrow)$ by $+\mu_B g_e B_N/2$ ($-\mu_B g_e B_N/2$). This allows measurement of B_N at $B_z > B_z^{cr}$ from the spectral splitting $\Delta E_{|+1\rangle,|-1\rangle} = E_{|+1\rangle} - E_{|-1\rangle}$ of the bright exciton doublet: $B_N \approx (\Delta E^0_{|+1\rangle,|-1\rangle} - \Delta E_{|+1\rangle,|-1\rangle})/\mu_B g_e$. $\Delta E^0_{|+1\rangle,|-1\rangle}$ is the bright exciton splitting at $B_N = 0$ that we deduce from pump-probe measurements where the sample is kept in the dark for a sufficiently long time to allow nuclear spins to relax (>200 s) [12]. Fig. 2 shows the dependence of $\Delta E_{|+1\rangle,|-1\rangle}$ and the corresponding value of B_N (right scale) on the intensity P_{exc} of circularly polarized light at B_z=6.0 T. Results for σ^+ (σ^-) excitation plotted with open (solid) symbols show the variation of $\Delta E_{|+1\rangle,|-1\rangle}$ with P_{exc}, and demonstrate the occurrence of nuclear spin pumping.

As can be inferred from Fig.2, there are two distinct regimes in the DNP. At low excitation power ($P_{exc} <$ 10 μW) positive $B_N > 0$ is induced for *both* σ^+ and σ^- polarized excitation. By contrast, at high powers ($P_{exc} > 10$ μW) $\sigma^{+(-)}$ excitation results in $B_N < 0$ ($B_N > 0$), similar to previous reports [1–5, 9]. We note however that such direct correspondence between the helicity of the excitation and direction of nuclear spin polarization is only observed in the regime of saturation and suppression of the exciton and biexciton PL which is accompanied by the occurrence of broad background emission in the PL spectrum. We thus conclude that the build-up of $|B_N|$ observed in this regime cannot originate from the HI with the ground state excitons in the QD. The most likely source of DNP in this case is the HI of the nuclei with the spin polarized electrons in the excited shells of the dot or in the wetting layer. This is further confirmed in an experiment with above GaInP barrier band-gap excitation: for a laser with photon energy $E_{exc} = 2.28$ eV high power DNP is suppressed almost completely, as electrons lose their spin polarization during energy relaxation. By contrast, the unusual polarization-insensitive DNP observed in the low power regime remains effective under this condition: independently of light polarization it leads to $B_N \approx +0.2$ T, similar to the case of $E_{exc} = 1.88$ eV.

As we show in more detail in the model presented below, these observations allow us to conclude that in the

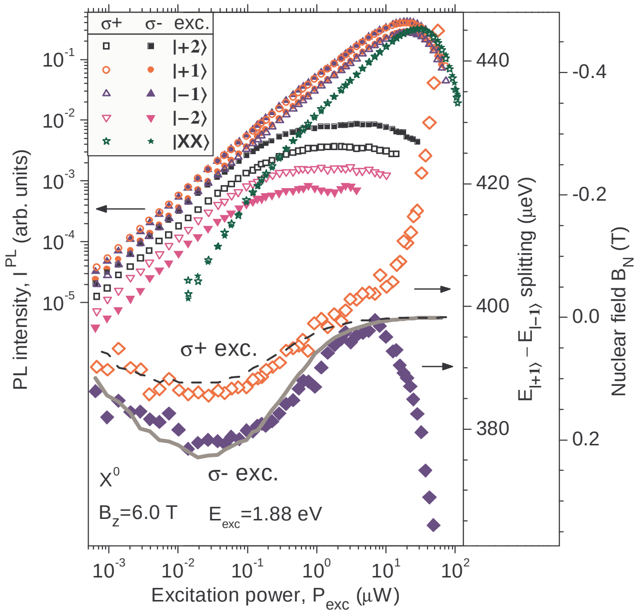

FIG. 2. Results of PL power dependence in a neutral dot at B_z=6 T under σ^+ (open symbols) and σ^- (solid symbols) excitation with photon energy 1.88 eV. PL intensities (left scale) for all four exciton states and for the total intensity of biexciton emission are plotted. Spectral splitting (right scale) of the two optically-allowed exciton states ($E_{|+1\rangle} - E_{|-1\rangle}$) is shown with diamonds. Lines show fitting obtained using the model presented in text. Additional scale on the right shows nuclear field B_N on the dot deduced from the splitting.

wide range of powers $P_{exc} < 10$ μW DNP is induced by virtual flips of the $|+2\rangle$ dark exciton into the intermediate $|+1\rangle$ state followed by recombination. This second order process dominates due to the asymmetry of the exciton spectrum seen in Fig.1: the energy splitting between $|+2\rangle$ and $|+1\rangle$ is significantly smaller than for $|-2\rangle$ and $|-1\rangle$ (see detailed explanation below). This results in $B_N > 0$ for both σ^+ and σ^- polarized excitation despite considerable differences in the exciton populations with $\uparrow$ and $\downarrow$ electrons. The interplay between processes responsible for nuclear spin pumping and depolarization results in the previously unobserved strongly non-monotonic power-dependence of B_N in Fig.2. The model presented below explains this behavior, and describes the underlying mechanisms quantitatively.

Transfer of spin polarization from the exciton to the nuclear system requires $e-N$ spin-flip that transforms a bright exciton into dark or vice versa [zigzag lines in Fig. 1 (c)]. The energy splittings between exciton states ($\sim$100 μeV) significantly exceed the nuclear spin level separation ($\sim$0.1 μeV) thus raising the problem of energy conservation [14]. The energy mismatch can be compensated by the photon emitted during recombination [1, 6, 10] in a second-order process. For example, if the QD contains a $|+2\rangle$ "dark" exciton it can make a virtual flip into the $|+1\rangle$ state increasing nuclear spin polarization by +1 [zigzag line Fig. 1 (c)]. At the second stage the "bright" $|+1\rangle$ exciton recombines with emission of a σ^+ photon [thick solid arrow in Fig. 1(c)]. Such processes which change nuclear spin polarization by $+1(-1)$ can start from $|+2\rangle$ or $|-1\rangle$ ($|+1\rangle$ or $|-2\rangle$) states [see Fig. 1(c)]. For each initial state only one intermediate state (with the same hole and opposite electron spin) is possible. Spin flips are governed by the off-diagonal matrix element $V_{hf} = \langle\Uparrow\uparrow| H_{hf} |\Uparrow\downarrow\rangle = \langle\Downarrow\uparrow| H_{hf} |\Downarrow\downarrow\rangle$ of the hyperfine Hamiltonian H_{hf} which is proportional to the in-plane component of the fluctuating nuclear field [15] and can be estimated as $|V_{hf}| \approx A_{hf}/(2\sqrt{N}) \sim 1$ μeV , where $A_{hf} \approx$230 μeV [16] is the electron spin splitting corresponding to fully polarized nuclear spins in the QD, and $N\sim 10^4 \div 10^5$ is the number of nuclei in the dot.

As PL peaks corresponding to $J_z \approx +2(-2)$ dark excitons are mainly $\sigma^+(\sigma^-)$ polarized the wave-functions of mixed states can be approximated as:

$$\begin{aligned} |\pm 2\rangle &= c_+ |\Uparrow\uparrow\rangle + s_+ |\Uparrow\downarrow\rangle \quad [c_- |\Downarrow\downarrow\rangle + s_- |\Downarrow\uparrow\rangle], \\ |\pm 1\rangle &= c_+ |\Uparrow\downarrow\rangle - s_+ |\Uparrow\uparrow\rangle \quad [c_- |\Downarrow\uparrow\rangle - s_- |\Downarrow\downarrow\rangle], \end{aligned} \quad (1)$$

with mixing parameters $c_\pm = \cos\phi_\pm$, $s_\pm = \sin\phi_\pm$ ($\phi_\pm \ll 1$).

The nuclear spin pumping rate is proportional to (i) the magnitude of the hyperfine mixing between the initial and intermediate states which is proportional to V_{hf}^2 and inversely proportional to the square of the energy splitting between them, (ii) the recombination rate of the intermediate exciton state $\cos^2\phi_\pm/\tau_r$ ($\sin^2\phi_\pm/\tau_r$) for $|\pm 1\rangle$ ($|\pm 2\rangle$) and (iii) the probability p_i to find the dot in the initial state i. The total spin pumping rate w_{pump} is a sum of the individual rates for all four possible initial states and can be calculated using wavefunctions from Eq.1 as:

$$\begin{aligned} w_{pump} = &\frac{\cos 2\phi_- V_{hf}^2}{\Delta E^2_{|-1\rangle,|-2\rangle}} \left(\frac{\sin^2\phi_-}{\tau_r} p_{|-1\rangle} - \frac{\cos^2\phi_-}{\tau_r} p_{|-2\rangle} \right) \\ &+ \frac{\cos 2\phi_+ V_{hf}^2}{\Delta E^2_{|+2\rangle,|+1\rangle}} \left(-\frac{\sin^2\phi_+}{\tau_r} p_{|+1\rangle} + \frac{\cos^2\phi_+}{\tau_r} p_{|+2\rangle} \right) \end{aligned} (2)$$

where $\Delta E_{|+2\rangle,|+1\rangle} = (E_{|+2\rangle} - E_{|+1\rangle})$, $\Delta E_{|-1\rangle,|-2\rangle} = (E_{|-1\rangle} - E_{|-2\rangle})$. The p_i are proportional to I_i^{PL}, the PL intensities plotted in Fig. 2, and inversely proportional to the photon emission rate:

$$\begin{aligned} p_{|\pm 2\rangle} &= (I^{PL}_{|\pm 2\rangle}/I_0) \times \tau_r \times \sin^{-2}\phi_\pm, \\ p_{|\pm 1\rangle} &= (I^{PL}_{|\pm 1\rangle}/I_0) \times \tau_r \times \cos^{-2}\phi_\pm, \end{aligned} \quad (3)$$

where the normalization constant I_0 corresponds to the detected PL intensity per one photon emitted by the dot.

For the splittings $\Delta E \lesssim 400$ μeV and $\tau_r \approx 1$ ns we can estimate $w_{pump} \gtrsim 10^4$ s^{-1} from Eq.2. For an alternative scenario involving phonon-mediated transitions between the "dark" and "bright" states the rate is below 10^1 s^{-1} [17] under similar conditions and can be neglected. It also follows from Eq.2 that the dark excitons play the major role in DNP. This is not only a result of their increased population at low P_{exc} [1, 18], but also a result of the high recombination rate $\tau_r^{-1}\cos^2\phi_\pm$ of the intermediate "bright" states. According to Eq.2 $|+2\rangle$ and $|-2\rangle$ excitons contribute to w_{pump} with opposite signs. However, for small $P_{exc} = 50$ nW at $B_z = 6.0$ T we detect $B_N \approx$+0.13 and +0.21 T corresponding to $\approx$5% and 10% nuclear spin polarization for σ^+ and σ^- excitation respectively. This is observed despite the substantial decrease (increase) of $|+2\rangle(|-2\rangle)$ population when the laser polarization is changed from σ^- to σ^+: $p_{|+2\rangle}/p_{|-2\rangle}$ is $\approx$2.6 times smaller for σ^+ excitation [see Fig.1(b)]. This indicates that DNP induced by the $|+2\rangle$ exciton is more efficient, which is explained by smaller energy splitting between the initial and intermediate states for the $|+2\rangle$ exciton: $\Delta E^2_{|+2\rangle,|+1\rangle} \approx \Delta E^2_{|-1\rangle,|-2\rangle}/4.5$ [see Fig.1(a)].

Nuclear spin depolarization induced by optical pumping can result from fluctuations of the electron Knight field [1] or electric field gradients interacting with nuclear quadrupole moments [13]. In our model we assume that recombination or capture of an exciton can result in simultaneous flip of a nucleus with probability R. In the low power regime ($P_{exc} < 10$ μW) the capture/recombination rate is proportional to the total PL intensity of all exciton and biexciton transitions I^{PL}_{tot}. Thus the optically induced depolarization rate can be approximated as:

$$w^{opt}_{dep} = 2R \times (I^{PL}_{tot}/I_0) \times (\mu_B g_e B_N/A_{hf}), \quad (4)$$

where I_0 is the same normalization constant as in Eq.3, and the factor of 2 arises from spin relaxation during

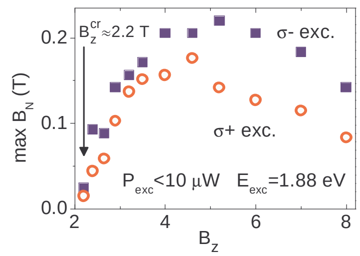

FIG. 3. Magnetic field dependence of the maximum nuclear field B_N induced at low power ($P_{exc} < 10\ \mu$W) of σ^+ (◯) and σ^- (■) polarized excitation with photon energy $E_{exc} =$ 1.88 eV.

capture and recombination. The non-optically induced relaxation rate (e. g. due to nuclear spin diffusion or coupling with phonons) can be approximated as

$$w_{dep}^{dark} = \tau_{dark}^{-1} \times (S_P + S_{In})/2 \times N \times (\mu_B g_e B_N / A_{hf})^2 \quad (5)$$

where $S_P = 1/2$, $S_{In} = 9/2$, and $\tau_{dark} \approx 200$ s is the nuclear spin decay time in the dark at $B_z = 6$ T [12].

In the steady-state condition $w_{pump} = w_{dep}^{opt} + w_{dep}^{dark}$. Using Eq.(2,3,4,5) we can explicitly express B_N as a function of the PL intensities I^{PL} measured in experiment. We note that when I_{tot}^{PL} reaches its maximum value the dot is saturated, which means that the sum of the probability to find the dot in the biexciton state $p_{|XX\rangle} = (I_{|XX\rangle}^{PL}/I_0) \times \tau_r/2$ and all probabilities in Eq. 3 is ≈ 1. This gives an additional relation that allows to eliminate I_0 from the final expression for B_N.

The calculated dependence $B_N(P_{exc})$ for $\sigma^{+(-)}$ polarized excitation is shown with a dashed (solid) line in Fig.2. The following magnitudes of the fitting parameters were obtained: $R{\approx}1{\cdot}10^{-3}$, mixing factors $\sin^2\phi_+{\approx}1.4{\cdot}10^{-2}$, $\sin^2\phi_-{\approx}1.0{\cdot}10^{-2}$, and $N{\approx}1.1{\cdot}10^4$. We also used τ_r=1 ns. The calculated curves are in good agreement with the experimental results for $P_{exc} <$ 10 μW. In particular the non-monotonic character in the low-power regime (with the B_N maximum at $P_{exc} \sim$50 nW and *depolarization* towards $P_{exc} \approx 1$ nW and $P_{exc} \approx 10\ \mu$W) is well reproduced and can now be explained as follows: At low powers $P_{exc} \approx 1$ nW, the rate w_{pump} is very small due to low exciton occupancy and non-optically induced relaxation with the rate w_{dep}^{dark} (see Eq.5) dominates. w_{dep}^{dark} remains constant with increasing P_{exc}, whereas the exciton occupancy increases leading to higher w_{pump}, and as a result a higher degree of nuclear polarization. At $P_{exc} > 50$ nW, the population of dark states gradually saturates leading to saturation of w_{pump}. On the other hand, the rate of optically induced depolarization (see Eq.4) increases linearly with power in a wide range of P_{exc}, and as a result nuclear polarization decreases.

Finally we consider the dependence of the maximum B_N induced in the low power regime as a function of $B_z > B_z^{cr}$ shown in Fig.3 for both helicities of below the GaInP barrier excitation. Both curves have flat maxima at $B_z \approx 5$ T. For higher fields, B_N decreases with B_z. This is consistent with the photon-assisted DNP interpretation due to the vanishing difference between the splittings $\Delta E^2_{|+2\rangle,|+1\rangle} \approx \Delta E^2_{|-1\rangle,|-2\rangle}$.

It can also be seen from Fig.3 that B_N is significantly reduced when B_z approaches the crossing field $B_z^{cr} \approx 2.2\ T$. This is a result of reduction of the w_{pump} rate (Eq. 2) due to anticrossing [11] of the $|+2\rangle$ and $|+1\rangle$ states, which originates from low-symmetry $e-h$ exchange interaction. It leads to a minimum splitting between the two corresponding PL lines $\delta_{exc}^e{\approx}$ 20 μeV at $B_z = B_z^{cr}$. In experiment the anticrossing manifests itself as a significant increase of the $|+2\rangle$ PL intensity as it gains oscillator strength from the $|+1\rangle$ state. The suppression of w_{pump} takes place in the vicinity of B_z^{cr} over a field range equal to several times the value of $\delta_{exc}^e/(\mu_B g_e)$. Anticrossing acts in several ways: (i) by setting the lower limit on $\Delta E_{|+2\rangle,|+1\rangle}{=}\delta_{exc}^e$ (see Eq.2), (ii) by reducing electron spin projections due to the non-hyperfine mixing of $|+2\rangle$ and $|+1\rangle$ states (corresponding to $\phi_+{=}\pi/4$ in Eq.1 at $B_z = B_z^{cr}$), and (iii) by reducing the population of the $|+2\rangle$ state due to increased emission probability. We thus conclude that DNP insensitive to light polarization should be enhanced for *symmetric* quantum dots where $\delta_{exc}^e \approx 0$.

In conclusion, we have demonstrated optical pumping of nuclear spins weakly dependent on the wavelength and polarization of the low intensity light. The phenomena have been shown to originate from intrinsic asymmetry in the energy spectrum of the excitonic energy levels. We note that this process may lead to *self-polarization* - the optically induced spontaneous alignment of nuclear spins in the limit of vanishing external field $B_z \approx 0$ predicted for neutral quantum dots in Ref. [18].

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